

# Electron–lattice kinetics of metals heated by ultrashort laser pulses

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We propose a kinetic model of transient nonequilibrium phenomena in metals exposed to ultrashort laser pulses when heated electrons affect the lattice through direct electron–phonon interaction. This model describes the destruction of a metal under intense laser pumping. We derive the system of equations for the metal, which consists of hot electrons and a cold lattice. Hot electrons are described with the help of the Boltzmann equation and equation of thermoconductivity. We use the equations of motion for lattice displacements with the electron force included. The lattice deformation is estimated immediately after the laser pulse up to the time of electron temperature relaxation. An estimate shows that the ablation regime can be achieved.

## I. INTRODUCTION

The first theoretical prediction of transient laser-induced nonequilibrium electron temperature phenomena in metals was made more than twenty years ago<sup>1</sup>. It was shown that an ultrashort laser pulse ( $\sim 10^{-13} - 10^{-12}$  s) produces a nonequilibrium state of the electron gas near a metal surface. However, experimental picosecond ( $\sim 10^{-12}$  s) laser studies of thermally assisted multiphonon photoemission were unable to measure, and even failed to observe, this nonequilibrium electron state<sup>2</sup>. This failure had a simple explanation in terms of the theory of electron–lattice thermal relaxation,<sup>3</sup> which yields a relaxation time of the order of  $\tau_{e-l} \sim 10^{-12}$  s. It was necessary to use power pulses shorter than  $\tau_{e-l}$ . Such measurements with subpicosecond ( $\sim 10^{-13}$  s) pulses revealed a transient nonequilibrium regime in transmittivity and IR reflection<sup>5–8</sup>, giant electron emission<sup>9–11</sup> and the emission of light<sup>12–14</sup>.

We briefly summarize the physical process. The ultrashort laser pulse ( $\Delta t \sim 10^{-14} - 10^{-13}$  s) absorbed in a metal raises the electron temperature  $T_e$  considerably higher than the lattice temperature because of the difference in their specific heats ( $c_e \ll c_l$ ). Subsequent electron cooling results mainly from two processes, namely electron–lattice thermal relaxation and electron thermoconductivity. These are usually modeled with a set of coupled thermoconductivity equations for the electron and lattice components. These equations are nonlinear and can generally be solved numerically, yielding the electron temperature relaxation. The solution also shows that the subsequent ablation regime can be achieved, which involves the "cold" destruction of a metal into the parts consisting of different phases. "Hot" destruction, namely melting, can also be studied with the help of this solution<sup>15</sup>. However, such an approach has several shortcomings. First, the question remains as to whether the equations of thermoconductivity are still hold at such high frequencies ( $\sim 1/\Delta t$ ). Second and more importantly, these equations can only describe the temperature dynamics of a metal but not electron transport, lattice deformation, thermionic emission, etc. It is evident that a strict kinetic approach is needed to describe the various transport phenomena properly and derive thoroughly the equation of thermoconductivity<sup>16</sup>.

In this paper we present a theory of transient nonequilibrium phenomena in metals subject to ultrashort laser pulses. Our theory is based on the Boltzmann equation for the nonequilibrium electronic partition function. We focus mainly on times shorter than the electron–lattice relaxation time  $\tau_{e-l}$ . Electrons therefore affect the lattice via direct electron–phonon interactions. To consider lattice deformations, we use the equations of the so-called dynamical theory of elasticity. Lattice deformation is due to the nonequilibrium electron state and results from the effective "driving" force (proportional to  $\nabla T_e^2$ ) on the lattice. This force also governs the renormalization (depending on  $T_e$ ) of the lattice constants (sound velocity and optical phonon gap). We show that the driving force leads to large lattice deformations, and can destroy the crystal. These results are in agreement with measurements of time-resolved X-ray diffraction synchronized with laser pumping<sup>17</sup>. A nonstationary increase in lattice parameters of Au(111) and Pt(111) single crystals was detected. Measurements of the shift and intensity variation of Bragg peaks, as well as the Debye–Waller factor, enables one to separate the effects of lattice deformation and heating. The transformation of elastic into plastic deformation was also observed.

The plan of the paper is as follows. In Sec. 2 we present the kinetic theory of the process under study: the Boltzmann equation for an electron gas and the elastic equation for the lattice are derived, along with the equation for thermoconductivity. In Sec. 3, the solutions of the proposed equations are found for the times of interest. The lattice deformation is calculated. In Sec. 4 the solutions are analyzed. The lattice deformation is estimated analytically in various limiting cases. The possibility of crystal destruction under laser pumping is discussed.

## II. THEORETICAL FRAMEWORK

Let us briefly recapitulate the main equations of our problem. For the lattice deformation we use the so-called equation of dynamical theory of elasticity<sup>18, 19</sup>

$$\rho \frac{\partial^2 u_i}{\partial t^2} - \lambda_{ijlm} \frac{\partial^2 u_l}{\partial x_j \partial x_m} = G_i, \quad (1)$$

where  $\rho$  is the lattice density,  $\lambda_{ijlm}$  is the tensor of elastic constants, and the driving force describes the effect of free carriers on the lattice

$$G_i = \frac{\partial}{\partial x_j} \int \frac{2d^3p}{(2\pi)^3} \lambda_{ij}(\mathbf{p}) f_p(\mathbf{r}, t). \quad (2)$$

The deformation potential  $\lambda_{ij}(\mathbf{p})$  yields the change in the local electron spectrum,

$$\delta\varepsilon(\mathbf{p}, \mathbf{r}, t) = \lambda_{ij}(\mathbf{p}) u_{ij}(\mathbf{r}, t).$$

To find the electron distribution function  $f_p(\mathbf{r}, t)$ , we use the Boltzmann equation with the electron-phonon collision integral

$$\begin{aligned} \text{St}f_p = & \sum_n \int \frac{d^3k}{(2\pi)^3} w_{pk}^{(n)} \delta(\varepsilon_{p+k} - \varepsilon_p + \omega_k^{(n)}) \left( (1 - f_p) f_{p'} N_k^{(n)} - f_p (1 - f_{p'}) (1 + N_k^{(n)}) \right) \\ & + \sum_n \int \frac{d^3k}{(2\pi)^3} w_{pk}^{(n)} \delta(\varepsilon_{p+k} - \varepsilon_p - \omega_k^{(n)}) \left( (1 - f_p) f_{p'} (1 + N_k^{(n)}) - f_p (1 - f_{p'}) N_k^{(n)} \right), \end{aligned} \quad (3)$$

with the probability of a scattering process involving a phonon of the  $n$ -th branch

$$w_{pk}^{(n)} = \frac{\pi}{\rho \omega_k^{(n)}} \left| e_i^{(n)} \lambda_{ij}(\mathbf{p}) k_j \right|^2,$$

where  $e_i^{(n)}$  and  $\omega_k^{(n)}$  are the polarization and spectrum of phonons of the  $n$ -th branch, respectively.

Since the phonon-phonon relaxation time is large ( $\sim 10^{-11}$  s) compared with the times of interest, the phonon distribution function  $N_k^{(n)}$  takes its equilibrium value at the lattice temperature  $T_\ell$ ,

$$N_k^{(n)}(T_\ell) = \frac{1}{\exp(\omega_k^{(n)}/T_\ell) - 1}.$$

The electron-electron relaxation time due to scattering on phonons  $\tau \sim T_\ell^{-1} \sim 10^{-14}$  s (see below) is much less than the characteristic time of laser pumping. Therefore the electron gas is nearly in thermal equilibrium at the local temperature  $T_e(\mathbf{r}, t)$ . We seek a solution of the Boltzmann equation in the form

$$f_p = f_0 \left( \frac{\varepsilon_p - \mu}{T_e} \right) + \chi_p \frac{\partial f_0}{\partial \varepsilon}, \quad (4)$$

where  $f_0$  is the local equilibrium Fermi-Dirac partition function and  $\chi_p$  is the nonequilibrium part. We obtain for the collision integral (3)

$$\text{St}f_p = \text{St}f_0 - \tau^{-1} \left( \chi_p - \frac{\langle \chi_p \rangle}{\langle 1 \rangle} \right) \frac{\partial f_0}{\partial \varepsilon}, \quad (5)$$

where the scattering rate

$$\tau^{-1} = T_\ell \sum_n \langle w_{pk}^{(n)} / \omega_k^{(n)} \rangle \sim \pi g^2 T_\ell.$$

The latter estimate is valid when the ion temperature  $T_\ell$  is considerably higher than the Debye temperature; the dimensionless electron-phonon coupling constant  $g \sim \lambda/\varepsilon_F \sim 1$ . The brackets denote integration over the Fermi surface

$$\langle \dots \rangle = \int \frac{2dS_F}{v(2\pi)^3} (\dots).$$

The first term in (5) comes from the contribution of the local equilibrium partition function:

$$\text{St}f_0 = \sum_n \int \frac{d^3p'}{(2\pi)^3} w_{pk}^{(n)} (f_0(\varepsilon_p) - f_0(\varepsilon_{p'})) \left( N_k^{(n)}(T_e) - N_k^{(n)}(T_\ell) \right) \times \left( \delta(\varepsilon_p - \varepsilon_{p'} - \omega_k^{(n)}) + \delta(\varepsilon_p - \varepsilon_{p'} + \omega_k^{(n)}) \right). \quad (6)$$

This term describes the energy flow from electrons to phonons when they are at different temperatures. This term is absent if the temperatures of the electron and lattice subsystems coincide.

The nonequilibrium part of the electron distribution function has to satisfy two conditions. The first is indeed the conservation law of the number of carriers:

$$\int \frac{d^3p}{(2\pi)^3} \chi_p \frac{\partial f_0}{\partial \varepsilon} = 0.$$

This expression determines the chemical potential and results in the renormalization of the deformation potential:  $\lambda(\mathbf{p}) \rightarrow \lambda(\mathbf{p}) - \langle \lambda(\mathbf{p}) \rangle / \langle 1 \rangle$ .

The second condition

$$\int \frac{d^3p}{(2\pi)^3} (\varepsilon_p - \mu) \chi_p \frac{\partial f_0}{\partial \varepsilon} = 0 \quad (7)$$

enables us to define the local temperature  $T_e$  (see Ref.<sup>20</sup>), i.e., to write the equation of thermoconductivity.

Substituting Eq. (4) into the Boltzmann equation, we get

$$\frac{\partial \chi_p}{\partial t} + \mathbf{v} \frac{\partial \chi_p}{\partial \mathbf{r}} + \frac{\chi_p - \langle \chi_p \rangle / \langle 1 \rangle}{\tau} = -e \mathbf{v} \mathbf{E} - \lambda_{ij}(\mathbf{p}) \frac{\partial u_{ij}}{\partial t} + \frac{\varepsilon_p - \mu}{T_e} \left( \frac{\partial T_e}{\partial t} + \mathbf{v} \frac{\partial T_e}{\partial \mathbf{r}} \right) + \text{St}f_0 \left/ \frac{\partial f_0}{\partial \varepsilon} \right. \quad (8)$$

To obtain the equation for the local temperature  $T_e(\mathbf{r}, t)$ , we multiply the Boltzmann equation (8) by  $(\varepsilon_p - \mu) \partial f_0 / \partial \varepsilon$  and integrate over  $\mathbf{p}$ . With the help of Eq. (7) we find the equation of thermoconductivity,

$$c_e(T_e) \frac{\partial T_e}{\partial t} + \text{div } \mathbf{q} = Q - \alpha(T_e - T_\ell), \quad (9)$$

where  $c_e(T_e) = \pi^2 \langle 1 \rangle T_e / 3 \equiv \beta T_e$  is the electron heat capacity and  $\mathbf{q}$  is the heat flow:

$$\mathbf{q} = \int \frac{2d^3p}{(2\pi)^3} \mathbf{v} (\varepsilon_p - \mu) \chi_p \frac{\partial f_0}{\partial \varepsilon}. \quad (10)$$

Using Eq. (6), we can find the last (relaxation) term on the right-hand side of Eq. (9). For high temperatures  $T_\ell, T_e \gg \omega_D$ , the electron-lattice relaxation constant  $\alpha$  is

$$\alpha = \sum_n \int \frac{2dS_F dS'_F}{vv'(2\pi)^6} w_{pk}^{(n)} \omega_k^{(n)}.$$

The density  $Q$  of laser energy absorbed by electrons can be taken in the form

$$Q(z, t) = I(t)(1 - R)\kappa e^{-\kappa z}, \quad (11)$$

where  $R$  is the reflection coefficient and  $\kappa$  is the inverse penetration depth. The function  $I(t)$  describes the pulse shape.

Eqs. (1) and (9) must be supplemented by the proper boundary conditions. We assume the simplest geometry: the metal occupies the half-space  $z < 0$ . Hence, the boundary conditions for the above equations are

$$\left. \frac{\partial T_e}{\partial z} \right|_{z=0} = 0, \quad \left. \frac{\partial u_z}{\partial z} \right|_{z=0} = 0, \quad (12)$$

signifying that the heat flow through the surface and the normal stress tensor component vanish on the surface.

We also need the boundary conditions for the kinetic equation (8) at the metal surface. These boundary conditions depend on the type of electron reflection from the surface. We assume the specular reflection for simplicity.

### III. DYNAMICS OF ELECTRON TEMPERATURE AND LATTICE DEFORMATION

The above equations are nonlinear and very complicated. However, it is possible to solve them in an important limiting case. Below we are interested in times shorter than the electron–lattice relaxation time  $\tau_{e-l} \sim c_e(T_e)/\alpha$ . In this case the lattice temperature can be set to the initial temperature  $T_0$ , and the last terms in Eqs. (8) and (9) can be omitted.

To solve the system (1), (8) for the half-space with the boundary condition (12), we use the even continuation of the temperature  $T_e(\mathbf{r}, t)$  and the partition function  $\chi_p$ , and the odd continuation of  $u_z(\mathbf{r}, t)$ , into the half-space  $z < 0$ :

$$T_e(z < 0) = T_e(-z, 0), \quad u_z(z < 0) = -u_z(-z, 0). \quad (13)$$

For the parallel components  $u_x$  and  $u_y$  one must use the even continuation, but owing to the fact that the external heat (11) depends only on  $z$ , these components are vanish. In Eq. (8) we discard  $\langle \chi_p \rangle$ , which represents the 'in-term' in the collision integral. This term accounts for carrier conservation, i.e., for the isotropic channel of collision processes. Therefore it does not affect the heat flow and lattice driving force.

The solution of Eq. (8) has the form

$$\chi_p(\mathbf{r}, t) = \int_{-\infty}^t dt' X_p(\mathbf{r} - \mathbf{v}(t - t'), t') \exp\left(-\frac{t - t'}{\tau}\right), \quad (14)$$

where  $X_p$  is the right-hand side of Eq. (8),

$$X_p = -\lambda_{ij}(\mathbf{p}) \frac{\partial u_{ij}}{\partial t} + \frac{\varepsilon_p - \mu}{T_e} \left( \frac{\partial T_e}{\partial t} + \mathbf{v} \frac{\partial T_e}{\partial \mathbf{r}} \right). \quad (15)$$

Substituting the solution (14), (15) into the heat flow (10) and integrating over the energy variable according to  $d^3p = d(\varepsilon_p - \mu)dS_F/v$ , we obtain

$$\mathbf{q}(\mathbf{r}, t) = -\frac{\pi^2}{6} \left\langle \int_{-\infty}^t dt' \exp\left(-\frac{t - t'}{\tau}\right) \mathbf{v} \left( \frac{\partial}{\partial t'} + \mathbf{v} \frac{\partial}{\partial \mathbf{r}} \right) T_e^2(\mathbf{r} - \mathbf{v}(t - t'), t') \right\rangle. \quad (16)$$

The expression (16) is linear in  $T_e^2$ . It is convenient to introduce the new function  $\Theta(\mathbf{r}, t) = T_e^2(\mathbf{r}, t)$  and take the Fourier transform with respect to space and time variables. Then Eq. (16) yields the Fourier component of the heat flow:

$$\mathbf{q}(\mathbf{k}, \omega) = \frac{i\pi^2}{6} \left\langle \frac{(\omega - \mathbf{v}\mathbf{k})\mathbf{v}}{\omega - \mathbf{v}\mathbf{k} + i\tau^{-1}} \right\rangle \Theta(\mathbf{k}, \omega). \quad (17)$$

Substituting this result into the equation of thermoconductivity (9), we obtain its Fourier component

$$-i\frac{\pi^2}{3} \left\langle \omega + \frac{(\omega - \mathbf{v}\mathbf{k})\mathbf{v}\mathbf{k}}{\omega - \mathbf{v}\mathbf{k} + i\tau^{-1}} \right\rangle \Theta(\mathbf{k}, \omega) = 2\kappa(1 - R)I(\omega)U(\mathbf{k}), \quad (18)$$

where  $I(\omega)$  is the Fourier transform of the pulse shape  $I(t)$ . The factor  $Q(\mathbf{k})$  describes the spatial distribution of the laser field (11),

$$U(k_z) = \frac{2\kappa}{\kappa^2 + k_z^2}$$

which depends only on  $k_z$ . Equation (18) yields the temperature dynamics of metals under laser heating with the time and space dispersion.

We now turn to the equation for lattice displacements (1). The driving force  $G_i(\mathbf{r}, t)$  can be evaluated as in the derivation of Eqs. (16) and (17). Both the local equilibrium partition function and nonequilibrium part (4) contribute to the integral (2). Expanding the integrals over the energy variable in powers of  $T_e/\varepsilon_F$  up to the second order, we obtain

$$G_i(\mathbf{k}, \omega) = -\frac{\pi^2}{6} \frac{\partial}{\partial \varepsilon_F} \left\langle \frac{\tau^{-1} \lambda_{ij}(\mathbf{p}) k_j}{\omega - \mathbf{v}\mathbf{k} + i\tau^{-1}} \right\rangle \Theta(\mathbf{k}, \omega). \quad (19)$$

In addition to the electron force (19) we also obtain the temperature-dependent renormalization of the elastic constants  $\lambda_{ijlm}$  (sound velocities) due to the interaction with electrons (electron loop in the phonon self-energy function). The dominant contribution in the range of interest comes from the local equilibrium partition function:

$$\lambda_{ijlm} \rightarrow \lambda_{ijlm} - \langle \lambda_{ij}(\mathbf{p}) \lambda_{lm}(\mathbf{p}) \rangle - \frac{\pi T_e^2}{6} \frac{\partial^2}{\partial \varepsilon_F^2} \langle \lambda_{ij}(\mathbf{p}) \lambda_{lm}(\mathbf{p}) \rangle.$$

The electron contribution to the sound velocity is second order in the electron temperature,  $\Delta s/s \sim T_e^2/\varepsilon_F^2$ .

Taking the Fourier transform of the left-hand side of Eq. (1), one needs to keep in mind the singularity at  $z = 0$  after continuation (13) of the function  $u_z$ . This singularity contributes the  $d\delta(z)/dz$ -term in the second derivative  $d^2 u_z/dz^2$ ; such a term accounts for surface effects. The Fourier transform with respect to the coordinate  $z$  over the entire space gives

$$-\rho(\omega^2 - s^2 k^2) u_z(k, \omega) = G_z(k, \omega) + k C(\omega), \quad (20)$$

where  $s = \lambda_{zzzz}/\rho$  is the longitudinal sound velocity in the  $z$ -direction. In the last term,  $C(\omega)$  must be determined from the boundary condition (12), and takes the form

$$C(\omega) = -2i\omega s \int \frac{dk}{2\pi} \frac{G_z(k, \omega)}{\omega^2 - s^2 k^2}. \quad (21)$$

We next proceed to the electron temperature and lattice deformations represented by Eqs. (18) and (20) in various limiting cases.

#### IV. SPATIAL VARIATION OF ELECTRON TEMPERATURE AND LATTICE DEFORMATION

Equation (18) describes the electron temperature evolution under ultrashort laser heating of metals. This equation generalizes the usual thermoconductivity equation<sup>1</sup>. We are interested in the wave vector  $k$ , which is the greater of the inverse skin depth  $\kappa$  ( $\sim 10^5 \text{ cm}^{-1}$ ) and the electron diffusion length  $v\sqrt{\tau t_0}$  during the laser pulse  $t_0$ . In the usual experimental situation  $\tau^{-1} \sim 10^{14} \text{ s}^{-1}$ ,  $\kappa v \sim 10^{13} \text{ s}^{-1}$ , and the hydrodynamic regime  $\kappa v \ll \tau^{-1}$  obtains. Thus, one can omit the term  $\mathbf{k}\mathbf{v}$  in the denominator of the left-hand side of Eq. (18). The dominant contribution comes from the diffusion pole  $\omega \sim \tau v^2 k^2 \ll \tau^{-1}$ . Therefore, we can also omit  $\omega$  everywhere in comparison with  $\tau^{-1}$  or  $\kappa v$ . The solution of the thermoconductivity equation reads

$$\Theta(z, t) = \Theta_0 + \frac{i}{\beta} \int_{-\infty}^{\infty} \frac{dk d\omega dt' dz'}{(2\pi)^2 (\omega + i D k^2)} I(t') e^{-i\omega(t-t') + ik(z-z') - \kappa|z'|}, \quad (22)$$

where the diffusion coefficient  $D = \tau \langle v_z^2 \rangle / \langle 1 \rangle$  is introduced. The constant  $\Theta_0 = T_0^2$  comes from the solution of the corresponding homogeneous equation, and represents the initial temperature. Evaluating the integral (22) with respect to  $\omega$  and  $k$ , we obtain

$$\Theta(z, t) = \Theta_0 + \int_{-\infty}^t dt' \int_{-\infty}^{\infty} dz' \frac{Q(|z'|, t')}{\beta \sqrt{\pi(t-t')D}} \exp\left(-\frac{(z-z')^2}{4(t-t')D}\right). \quad (23)$$

We see immediately that the function (23) satisfies the boundary condition (12). For the temperature at the surface  $z = 0$ , Eq. (23) gives

$$T_e^2(0, t) = T_0^2 + \frac{4}{\pi\beta} \int_0^t dt' Q(0, t-t') e^{\kappa^2 D t'} \text{erfc}\left(\sqrt{\kappa^2 D t'}\right).$$

The electron temperature (23) just after the pulse peaks at the surface:

$$T_{max}^2 \sim \frac{I t_0 (1-R)}{\beta} \min\left(\kappa, (D t_0)^{-1/2}\right). \quad (24)$$

This result has a simple explanation. For short pulses  $\kappa\sqrt{Dt_0} \ll 1$  the time dependence of the temperature corresponds to the local laser intensity at the observation point. In the opposite case,  $\kappa\sqrt{Dt_0} \gg 1$ , the temperature distribution is determined mainly by the diffusion process.

Consider now the equation for lattice displacements (20) with the force (19). Note that in the hydrodynamic regime,  $\kappa v \ll \tau^{-1}$ , the dominant contribution to the force  $G_i$  comes from the local equilibrium partition function, i.e., the first term in (4), if we consider times greater than the electron–electron relaxation time,  $t \gg \tau$ . In this case, the force has the simple expression

$$G_i(\mathbf{r}, t) = \Lambda_{ij} \frac{\partial T_e^2(\mathbf{r}, t)}{\partial x_j},$$

where the constants

$$\Lambda_{ij} = \frac{1}{32\pi} \frac{\partial}{\partial \varepsilon_F} \int \frac{dS}{v} \lambda_{ij}(\mathbf{p}) \sim g\beta$$

are of the order of the electron density of states at the Fermi surface.

From Eq. (20) with the help of the expression (21) one can find the lattice deformation

$$\frac{du_z}{dz} = \frac{i\Lambda_{zz}\kappa(1-R)}{\rho\beta} \int \frac{d\omega dk}{(2\pi)^2} \frac{k^2 U(k) I(\omega)}{(\omega + ik^2 D)(\omega^2 - s^2 k^2)} \left[ e^{ikz} - e^{i\omega|z|/s} \right] e^{-i\omega t}. \quad (25)$$

The first term in the brackets in Eq. (25) represents the particular solution of the inhomogeneous Eq. (1) while the second corresponds to the general solution of the homogeneous form of Eq. (1), and represents the effect of the surface. The integrand in (25) contains poles associated with the diffusion and sound-wave excitations. Sound singularities are bypassed using infinitesimal phonon damping,  $\omega \rightarrow \omega + i0$ .

## V. EFFECT OF ACOUSTIC AND OPTICAL DISPLACEMENTS ON DESTRUCTION OF METALS

Equation (25) describes the effect of nonequilibrium electron heating on lattice deformations of acoustic type. This deformation vanishes at the surface  $z = 0$  according to the boundary condition (12). For  $z \neq 0$ , the second term in brackets in (25) represents a deformation wave propagating from the surface into the bulk of the metal. It makes a nonzero contribution only at sufficiently small depths  $z < st \sim 10^{-7}\text{cm}$ . Thus, we see that the deformation (25) peaks at  $z \sim 10^{-7}\text{cm} \ll \kappa^{-1}$ . To obtain the order of the effect, we can drop the second term in parentheses. It is then convenient to integrate over  $\omega$ , substituting the Fourier transform  $I(\omega)$ . We obtain

$$\frac{du_z}{dz} \sim \frac{\Lambda_{zz}\kappa(1-R)}{\rho\beta} \int_0^t dt' I(t') \int \frac{dk}{2\pi} U(k) \left( \frac{e^{-isk(t-t')}}{s(s+ikD)} - \frac{e^{-k^2 D(t-t')}}{k^2 D^2 + s^2} \right) e^{ikz}. \quad (26)$$

Consider times greater than the duration of a pulse  $t > t_0$  but less than the characteristic time of electron diffusion ( $t < (\kappa^2 D)^{-1} \sim 10^{-12}\text{s}$ ) and a sound-wave period ( $t \ll (s\kappa)^{-1} \sim 10^{-11}\text{s}$ ) with characteristic wave vector of the order of the inverse skin depth  $\kappa$ . In this range we can expand the exponentials in (26) in powers of  $t$  up to second order:

$$\frac{du_z}{dz} \sim \frac{\Lambda I t_0 \kappa (1-R)}{2\rho\beta} t^2 \int_{-\infty}^{\infty} \frac{dk}{2\pi} U(k) k^2 e^{ikz}. \quad (27)$$

Using the estimate  $\Lambda/\rho \sim gs^2/\varepsilon_F^2$  and the Eq. (24) for the maximum electron temperature, we extrapolate our result up to electron diffusion times  $t \sim (\kappa^2 D)^{-1}$ :

$$\frac{du_z}{dz} \sim g(1-R) \frac{It_0}{\kappa\beta} \left( \frac{s}{\tau v^2 \varepsilon_F} \right)^2 \sim g \left( \frac{sT_{max}}{\kappa \tau v^2 \varepsilon_F} \right)^2. \quad (28)$$

Setting  $s/v \sim 10^{-2}$ ,  $\kappa \sim 10^5 \text{ cm}^{-1}$ , we arrive at the numerical estimate  $du_z/dz \sim 10^{-2} g^5 (T_e/\varepsilon_F)^2$ .

Our result contains the natural factor  $T^2/\varepsilon_F^2$ , which means that laser heating is important as soon as the electron temperature is higher than the Fermi energy. Although the estimate was obtained for  $T_e \ll \varepsilon_F$ , it is still roughly correct up to  $T_e \sim \varepsilon_F$ . The additional small factor  $s^2/(v\kappa l)^2$  is due to the fact that the characteristic period of the

sound wave ( $10^{-11}s$ ) is much greater than the characteristic times of electron diffusion ( $10^{-12}s$ ) and laser heating ( $10^{-13}s$ ). Therefore it would be of considerable interest to calculate the lattice deformation from high-frequency (but long-wavelength) excitations, i.e., optical phonons whose period is about  $10^{-14}s$ . This case differs from the calculations above in the equation of lattice motion (1) and electron force (2), due to the different form of deformation potential (see<sup>21</sup>). Estimates show that the relative optical displacement (with respect to the lattice constant) are of order  $T^2/\varepsilon_F^2$ .

## VI. CONCLUSIONS

Our result for acoustic deformation (28) agrees with the experiment reported by P. Rentzepis [17] where a deformation  $du_z/dz \sim 10^{-3}$  had been observed in the laser heating of noble metals. However, we see that the interaction of heated electrons with optical phonons can provide a more effective means of strong lattice deformation, but this case has yet to be studied experimentally. An ultrashort intense laser pulse can result in the destruction and ablation of metals, while only the electron component is heated, and the lattice stays at considerably low temperature.

In conclusion, we emphasize two points. First, as follows from Eq. (9), the driving force for lattice expansion is proportional to  $T_e \partial T_e / \partial z$ . Because of the high absorption coefficient of metals in the UV ( $\kappa \sim 10^5 \text{cm}^{-1}$ ), the temperature gradient reaches  $\sim 10^9 \text{K/cm}$ . Note that the extremely high values of this parameter (which is typical of metals) leads to nonequilibrium expansion of the lattice. Second, subpicosecond elastic deformation of the lattice of order  $10^{-3} - 10^{-2}$ , corresponding to an internal pressure  $10 - 100 \text{GPa}$ , can provide an effective mechanism for subsequent laser fracture of metals.

We are grateful to S. I. Anisimov and V. A. Benderskii for many useful discussions and valuable comments. This work was supported in part by the Russian Foundation for Basic Research, Grant No. 97-02-16044. One of the authors (EGM) also thanks KFA Forschungszentrum, Jülich, Germany, for a Landau Postdoctoral Fellowship.

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